Heat Capacity Characterization of a 4 K Regenerator with non-Rare Earth Material

W. Chen, M. Jaeger, K. Rule

Creare LLC
Hanover, NH 03755

ABSTRACT

This paper presents separate effects test results of an advanced regenerator for a 4 K regenerative cryocooler. The regenerator is made of a non-rare earth material. It utilizes a unique approach to significantly enhance the regenerator's thermal capacity, which is the key to the efficient operation of a regenerative cryocooler. The regenerator employs a unique microchannel configuration that enables it to achieve high thermal performance at a low pressure drop. These performance features together will enable a regenerative cryocooler to achieve high thermal efficiency at low temperatures. To determine its effective heat capacity and heat transfer performance under prototypical conditions, the thermal response of the regenerator to a step temperature change in the inlet flow is characterized. This paper first describes the testing approach and the test setup. It then presents the measured heat capacity and the predicted values. Finally, the volumetric specific heat of the regenerator is compared with those of typical rare earth materials used in a 4 K regenerator.

INTRODUCTION

Superconductor digital electronics utilizing Josephson Junctions have very fast switching time and are able to operate at very high speeds in excess of 30 GHz for wideband, high frequency RF signal processing. A critical need for these superconductor electronics is reliable, efficient, and portable cryocoolers that can provide cooling at temperatures below 4 K to maintain the superconductivity of niobium-based microelectronics. Pulse tube cryocoolers are a promising technology for this application because there are no moving parts at their cold ends, minimizing mechanical vibration transmitted to cooled sensors and electronics and enhancing reliability. However, it is currently not possible for this type of cryocooler to reach temperatures below 4 K while maintaining acceptable thermal efficiency. The poor performance is mainly caused by the performance degradation in the regenerator at very low temperatures.

The losses in the regenerator account for more than 50% of the losses in a typical 4 K pulse tube cryocooler (Radebaugh et al. 2009). At low temperatures, one of the performance-limiting factors for a regenerator is its very limited thermal capacity. The specific heat of most conventional regenerator matrix materials falls off sharply with decreasing temperatures below about 40 K, proportional to the cube of the temperature ($Cp \propto T^3$). The decrease in specific heat substantially reduces the thermal mass of the regenerator at the cold end, limiting its capacity to cool the gas flowing to the cold expansion space. The low thermal capacity in the regenerator

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causes a large temperature swing during periodic heat transfer and significantly reduces the efficiency of the regenerator.

The problem of limited thermal capacity in low-temperature regenerators cannot be solved by simply increasing the regenerator size. Increasing the size of a regenerator can indeed increase its heat transfer area and heat capacity. However, these benefits can be offset by the penalties associated with a larger regenerator void volume. A larger void volume increases compression heating and expansion cooling associated with the pressure oscillation in the regenerator. It also increases the gas flow rate at the warm end of the regenerator because of a larger dead volume.

Current research efforts have focused on utilizing rare-earth compounds that have relatively large magnetic heat capacities at low temperatures in the vicinity of their magnetic ordering transition temperatures. These compounds, however, are either very brittle (e.g., rare-earth/transition metal based intermetallic compounds) or very chemically reactive, limiting their practical use (Pecharsky et al. 1997). In addition, it is very difficult to use rare-earth compounds to produce small uniform spheres for packed beds or mesh screen with thin wires for a plate stack (Mérida and Barclay 1998). Furthermore, the peak heat capacity of each magnetic material only spans a very narrow temperature range. A hybrid matrix with layers of materials having different transition temperatures is required to achieve adequate heat capacity for sufficient performance.

REGENERATOR WITH ADSORBED HELIUM TO ENHANCE HEAT CAPACITY

The high heat capacity of the process helium in a pulse-tube cryocooler is the main reason why it is very challenging for a regenerator to achieve a high thermal efficiency. But the high heat capacity can also be the answer to the problem: adsorbed (trapped) helium can be used to store heat from the process gas (helium) passing through the regenerator. Porous adsorbents with high porosities, such as activated carbon, can adsorb significant amounts of helium at low temperatures. Moreover, the specific heat of helium molecules trapped in proper adsorbents remains very high. Consequently, the effective volumetric specific heat of helium in an adsorbent can be higher than those of rare-earth compounds (such as ErPr, GdRh, and ErNi) currently used in low-temperature regenerators. The specific heat of adsorbed helium and its macroscopic density in an adsorbent are discussed below.

Specific Heat of Adsorbed Helium Is Very High

The specific heat of helium depends strongly on the pore geometry and can be very high. The heat capacity of ^{4}He confined in adsorbents, such as molecular sieves, increases linearly with temperature (T) in the range between 3.25 K and 9 K (Wada et al. 1984). The adsorbed helium specific heat at temperature T can then be approximated as $(2.2 \times T)$ J/mol-K for $3.5 < T < 9$ K (Wada et al. 1984). It was also found that the specific heat of adsorbed helium can decrease as the channel dimensionality and pore size in the adsorbent material decrease (Konishi et al. 1993). The specific heat of helium molecules in an adsorbent can also decrease as the helium concentration in the adsorbent increases. Our study suggests that the minimum pore size of an optimum adsorbent should be larger than 20 Å (i.e., mesoporous adsorbents) to prevent a potential decrease in helium specific heat due to reduction in dimensionality.

Helium Densities in Adsorbents Can Be Very High

Typically, the macroscopic density of helium in an adsorbent is controlled by the porosity, pore size distribution of the adsorbent, and ambient pressure. For this particular application, the ambient helium pressure is very high, about 1 MPa in a typical pulse-tube cryocooler. This pressure is significantly higher than the capillary pressure associated with the porous structure. Consequently, the adsorbed helium densities in mesoporous adsorbents (20 Å < pore size < 500 Å) should be close to that in microporous adsorbents (pore size < 20 Å). Using a mesoporous adsorbent prevents a potential decrease in helium specific heat. For these reasons, the helium adsorption

density is largely controlled by the porosity of the adsorbent. Activated carbon can achieve a porosity higher than 60%, and a concentration of adsorbed helium higher than 0.1 g/cm^3 at 10 K (Duband et al. 1988).

MICROCHANNEL REGENERATOR WITH HELIUM ADSORBENT

Even though activated carbon can achieve very high helium adsorption capacities, its thermal conductivity is very low at temperatures below 20 K. This makes it difficult to transfer heat in or out of the helium/activated carbon mixture at high frequencies. Furthermore, thermally coupling activated carbon powders to a heat transfer surface is quite challenging. Due to these challenges, previous efforts to incorporate helium adsorbents into low-temperature regenerators have met with very limited success (Jones et al. 1987; Pron'ko et al. 1976). To overcome these problems, we developed a unique regenerator design with helium adsorbent in intimate thermal contact with convective transfer surfaces via a heat spreader. The helium adsorbent is a monolithic adsorbent directly integrated into the heat spreader surface by introducing micro pores in the heat spreader substrates. Consequently, there is no thermal resistance or thermal stresses at the interface between the adsorbent and the heat spreader. This approach also eliminates potential particulate contamination in the regenerator because the adsorbent is not a collection of loose particles.

The regenerator core consists of a stack of thin, micromachined silicon disks separated by thin polymer thermal insulators (Fig. 1). Each silicon disk has a thick layer of monolithic helium adsorbent at the center and an annular array of slots/fins farther out. The single crystal silicon disks serve as a heat spreader to effectively transfer heat from the process gas to the adsorbed helium in the helium adsorbent. When gas passes through the regenerator, thermal energy from the process gas first transfers to the fins near the outer rim of the disk, then spreads radially inward to the center base plate holding the adsorbent, and finally moves axially into the adsorbent, where a large amount of adsorbed helium provides a high heat capacity. Because the thermal diffusivity of silicon at low temperature is very high, thermal waves can easily pass from the fins to the center base plate.

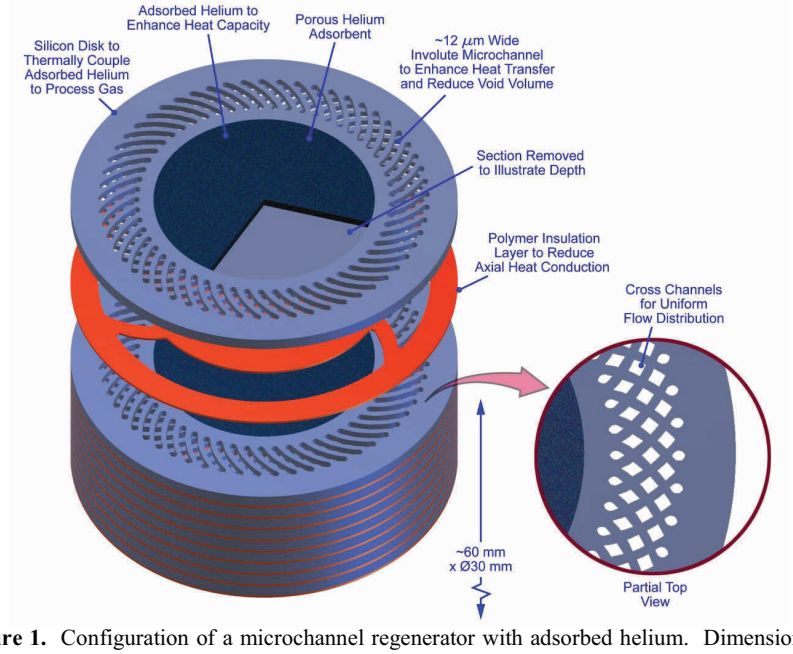
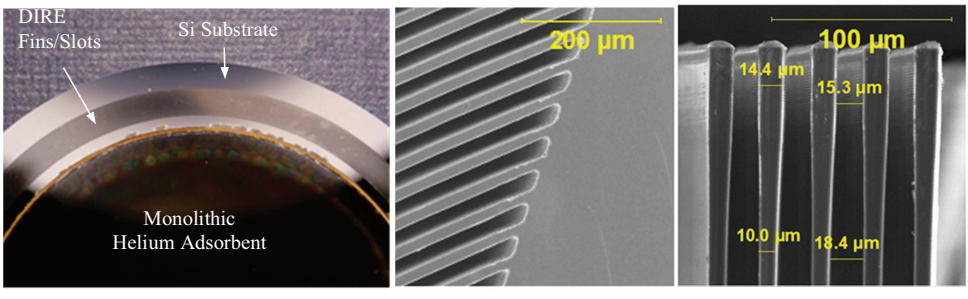


Figure 1. Configuration of a microchannel regenerator with adsorbed helium. Dimensions are not to scale for clarity.

Microchannels are machined in the silicon disks to form small flow paths for process helium, as shown in Fig. 2. Hundreds of involute slots are machined through the thickness of the wafer by Deep Reactive Ion Etching (DRIE). The involute spiral direction on one side of a disk is opposite to that on the other side. This allows the process gas to redistribute and helps to ensure that the flow distribution in the core is not sensitive to small variations in the channel geometry. The polymer spacer on each silicon disk reduces the conductive heat transfer along the regenerator axis. The composite disks are stacked together to form the regenerator core.

Brunauer-Emmett-Teller (BET) adsorption analysis shows that the pore surface area in the adsorbent is roughly 998 m^2/g, which is extremely high. The pore size distribution is shown in Fig. 3, suggesting the average pore diameter is roughly 3.5 nm, which is within the targeted range of 3 to 5 nm for this application.

The entire stack is housed inside a thin-walled titanium alloy shell. The stack is sandwiched between the hot end and cold flow distributors. One of the distributors includes a preload spring that maintains the composite disks in compression. The preload spring prevents the plates in the stack from separating from each other by gravity or vibrations and compensates for the mismatch in thermal expansion between the shell and the core.



(a) (b) (c)

Figure 2. Regenerator disks: (a) fin area of the silicon plate; (b) close-up view of the fin areal; (c) fracture surfaces of slot/fin features.

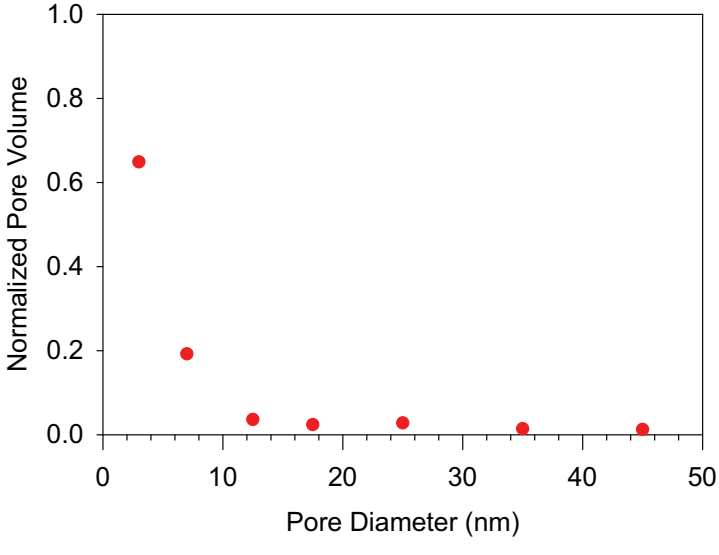


Figure 3. Pore size distribution for Creare's monolithic adsorbent.

OPERATION OF A HELIUM ADSORBENT REGENERATOR IN A CRYOCOOLER

The operation of a helium adsorbent regenerator in a pulse-tube cryocooler is very similar to a conventional regenerator. The main difference is during the cooldown process. In a cryocooler with a helium adsorbent regenerator, an appreciable amount of helium in the cryocooler will be adsorbed into the adsorbent during the cooldown process. This will cause the mean system pressure to decrease if no supplemental helium is supplied. This issue can be easily mitigated by slightly increasing the size of the helium reservoir or the cryocooler charge pressure to maintain its operating pressure after its cooldown. The helium adsorption during the cooldown process will also release additional adsorption heat into the regenerator, and thus increase the cooldown time. The estimated additional cooldown time is on the order of one minute. During steady state operation, however, there will be no noticeable difference between the operation of a helium adsorbent regenerator and a conventional regenerator.

The cycle period of pressure waves in a portable low-temperature pulse-tube cryocooler is very short, only about 50 ms (i.e., about 20 Hz). This cycle period is several orders of magnitude shorter than the adsorption and desorption time constant in an adsorbent, which is typically on the order of tens of seconds. Furthermore, the flow resistance between the adsorbent and process gas is quite high. Therefore, there will be no dynamic adsorption or desorption of helium in the adsorbents due to pressure oscillation or small local temperature swing in the regenerator within a pressure cycle. Here, it is the sensible energy of the adsorbed helium molecules being utilized, not the latent energy associated with adsorption and desorption.

REGENERATOR HEAT CAPACITY MEASUREMENT METHOD

We used a transient test technique to measure the regenerator heat capacity. We exposed the regenerator matrix to a step change in the inlet gas temperature and measured the temperature at the inlet and outlet as the thermal wave passed through the regenerator. The heat capacity is determined by the temporal profile of gas flow temperature at the inlet and outlet of the regenerator. The expected gas temperature at the regenerator outlet is illustrated in Fig. 4 when the gas inlet temperature has a "step" increase. The breakthrough time and the temporal profile of the outlet temperature are a function of the regenerator heat capacity, the flow heat capacity rate, and the thermal conductance between the gas flow and the matrix. Assuming a constant specific heat for the helium flow, the product of the cross-hatched area between the two temperature profile curves (Fig. 4) and flow heat capacity rate $\dot{m}c_p$ is the energy absorbed by the regenerator. With the helium gas flow rate held constant, the regenerator heat capacity is proportional to the break through time, $\Delta t_{breakthrough}$.

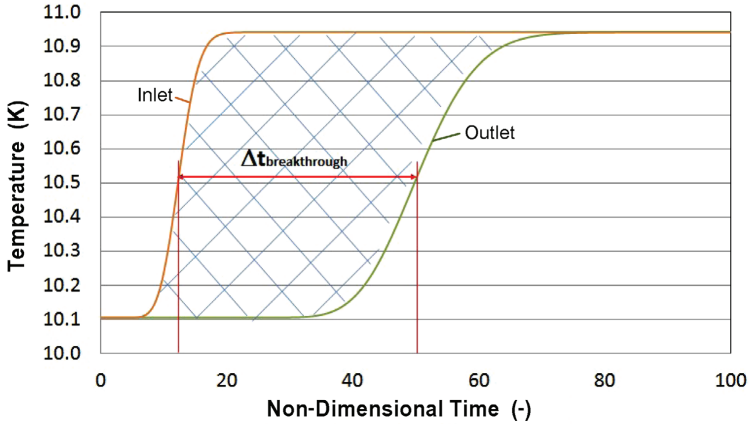


Figure 4. Illustration of gas temperature profiles at regenerator inlet and outlet as a function of time as thermal wave passes through regenerator.

The gas flow rate in this series of tests was selected so that the thermal wave breakthrough time is on the order of several seconds. This breakthrough time was long enough for our data acquisition system to resolve, but short enough to minimize the effects of parasitic heat leaks.

This test method was verified previously with a regenerator consisting of a stack of sintered stainless steel sheets.

TEST FACILITY

Fig. 5 shows a schematic of the test facility. Helium from a research-grade gas cylinder first passed through a mass flow meter, a gas cleaning subsystem (not shown) to remove contaminants, and then through a coil submerged in LN_2. It was cooled to about 100 K at the outlet of the coil. Next, it passed through a cold plate attached to a GM cold head to cool down to about 30 K and then through a Sumitomo cryocooler to further reduce its temperature to about 10 K. Finally, it passed through a heater and then entered the regenerator. At the beginning of each test, the heater was disabled and the entire regenerator was cooled down to a temperature very close to the temperature setpoint of the Sumitomo cold head. Once the regenerator temperature reached a steady state, the thin-film heater was turned on to rapidly raise the temperature of the helium flow entering the regenerator by 2 to 3 K. After the thermal wave passed through the regenerator, the heater was turned off, introducing another thermal wave through the regenerator, which cooled down the regenerator by 2 to 3 K back to the Stage 2 cold head temperature.

To reduce parasitic heating into the regenerator, we placed a cooling shroud around the regenerator. The cooling shroud was cooled by the exhaust flow from the regenerator. A relatively high-speed data sampling rate of 100 Hz was used to measure and record the transient gas flow temperatures at the inlet and outlet of the regenerator. We conducted a series of tests with regenerator inlet temperatures ranging from 10 to 22 K. The minimum test temperature was limited by the cooling capacity of the Stage 2 cold head. The heat load on the cold head was high because the test facility did not have a recuperator to reduce the heat load on the cold head. Tests were conducted at two different pressures to assess effects of the ambient helium pressure on the effective heat capacity of the adsorbed helium. The maximum pressure was limited to about 1.6 bar to limit the amount of helium in the regenerator dead volume.

Fig. 6 shows the regenerator sample used for characterizing the heat capacity of adsorbed helium/helium adsorbent. The regenerator consists of 80 plates with Kapton insulators in between the regenerator disks to reduce axial conduction. To minimize the effects of parasitic thermal mass, we located Swagelok fittings for temperature sensors, which have a relatively large thermal mass, at the two ends of the test apparatus where the fittings were not directly in the gas flow path. We also fabricated two thin sheathed Cernox sensors and used them to directly measure the gas flow temperatures at the inlet and outlet of the test stack to reduce the measurement response time. The Cernox sensors were located less than 0.25 in. away from the ends of the regenerator core.

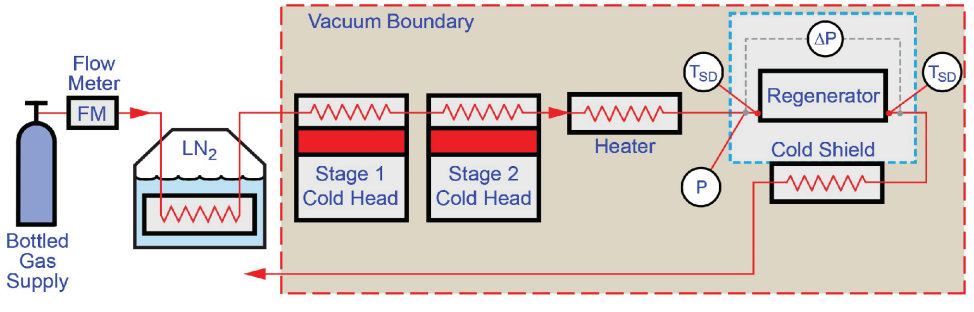
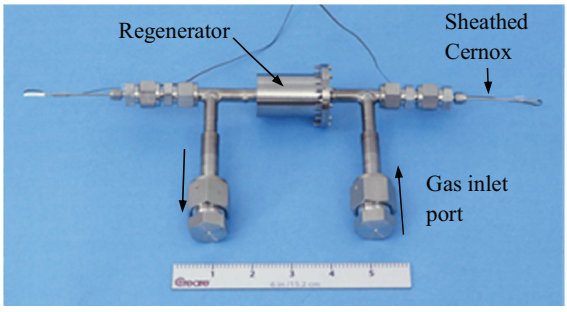
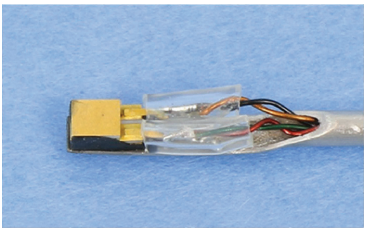


Figure 5. Test setup to characterize subscale regenerator heat capacity and thermal performance.





Sheathed SD Cernox with sensor exposed to gas flow

Figure 6. Regenerator sample with sheathed temperature sensors.

The regenerator test sample was designed to minimize the parasitic thermal mass for the core, which included the entire housing and manifold plates, as well as the dead volume inside the regenerator. At the nominal test temperature of 10 K, the resulting parasitic thermal mass was less than 10% of the core thermal mass, even if the entire parasitic mass participated (i.e., the thermal wave penetrated into all components and their temperatures all rose to the final temperature). The resulting dead volume was small enough that the resulting delay in thermal breakthrough was less than 2% of the actual breakthrough time.

DATA PROCESSING

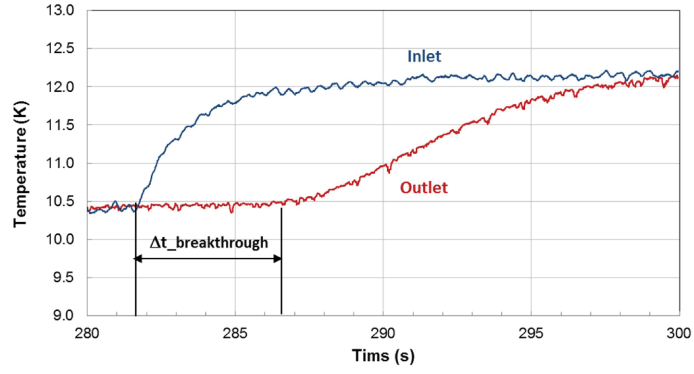
Fig. 7 shows representative measured temperature profiles of helium flow entering and leaving the regenerator core during thermal wave breakthrough tests. Because of the participating parasitic thermal mass in the heater and the transfer line between the heater and the core, the temperature of the inlet gas took a few seconds to reach its steady state value. As the gas temperature started to rise rapidly at t = 281.7 s, the temperature of the helium exiting the core remained unchanged because the thermal wave had not reached the outlet of the stack. After t = 286.5 s, the outlet temperature began to rise, but at a rate slower than that of the inlet flow.

Because the thermal conductance (UA) of each plate was very high relative to the helium flow rate of 35 SLPM, the thermal effectiveness of each regenerator plate was very close to 1 (i.e., each plate had an NTU value close to 1). Therefore, the slow temperature rise after thermal breakthrough at the outlet flow was not caused by the limited heat transfer between the helium flow and the plates in the core. Two possible causes for the slow outlet flow temperature rise are the following:

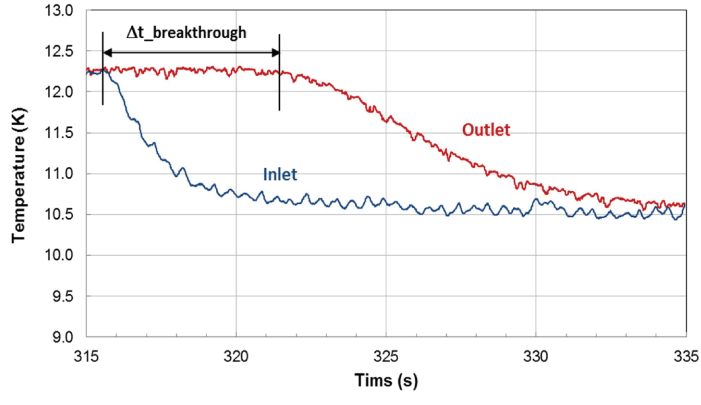
1) The parasitic thermal mass of the inlet and outlet headers and other parts that are thermally connected to the headers. Because of the limited convective heat transfer between the helium flow and headers, very little parasitic thermal mass was participating at the thermal wave front. Therefore, the parasitic thermal mass did not delay the initial thermal breakthrough at the outlet, but only broadened the wave front.

2) Helium desorption associated with the temperature increase in the adsorbent. For these separate effects tests, the thermal wave breakthrough time was about 5 to10 s, which was long enough that a small fraction of adsorbed gas could be desorbed. The associated desorption heat could artificially increase the effective heat capability of the test core. The associated desorption heat would also broaden the thermal wave front (i.e., the active heat transfer zone) in the regenerator. However, it should not have had appreciable effect on the initial breakthrough time since desorption does not occur right at the wave front.

To exclude the effects of parasitic thermal mass and the desorption heat from the regenerator core heat capacity from the measurement, the initial thermal breakthrough time was used to estimate the core heat capacity, which was taken as the product of the helium flow heat capacity



(a) Thermal Reponse with Increasing Inlet Temperature



(b) Thermal Reponse with Decreasing Inlet Temperature

Figure 7. Measured regenerator inlet and outlet temperature profiles.

rate and initial breakthrough time. The initial breakthrough time was taken as the time difference for the temperatures of the outlet and inlet flows to change by 2.5% of the total temperature rise, as shown in Fig. 7.

TEST RESULTS AND DISCUSSIONS

Fig. 8 shows the measured volumetric specific heat of adsorbent/adsorbed helium composite based on the data processing approach discussed above. The measured volumetric specific heat values are comparable with those of typical rare earth materials used in low temperature regenerators. The volumetric specific heat remains very high in the entire test temperature range from 10 to 22 K. This is in contrast to the specific heat of a rare-earth material, which typically peaks over a narrow temperature range of a few Kelvins. The wide temperature span of high specific heat allows the use of the adsorbent/adsorbed helium to serve as the thermal storage media over the entire temperature range below 20 K.

The measured specific heat increases slightly when the ambient helium pressure is increased from 1.0 bar to 1.6 bar. It is worth noting that the helium pressures in these tests are significantly lower than typical mean pressures in low temperature cryocoolers, which are typically about 10 bar. Test data for helium adsorption in activated carbon at temperature above 10 K indicates that specific helium adsorption capacity still increases appreciably with the ambient pressure until the pressure is significantly higher than 20 bar. This suggests the volumetric specific heat of adsorbed helium in actual applications can be even higher than the values shown here.

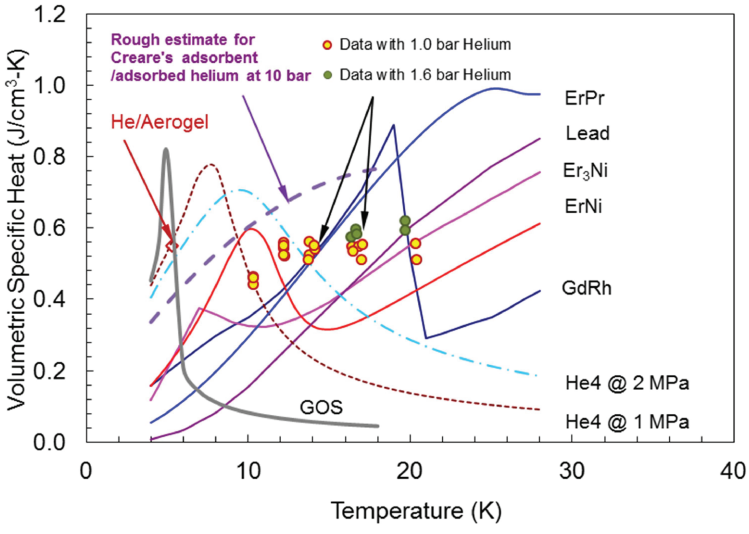


Figure 8. Comparison of measured volumetric specific heat of adsorbent/adsorbed helium with typical rare earth materials for low-temperature regenerators.

The measured specific heat is consistent with rough estimate values (Fig. 8). The volumetric specific heat of adsorbed helium is not well established in published literature. Its values are adsorbent dependent, and experimental data are scarcely available. To roughly estimate these values, the empirical correlation for adsorbed helium specific heat in Y zeolite, $(2.2 \times T)$ J/mol-K for $3.5 < T < 9$ K, was used (Wada et al. 1984). The adsorbed helium density was taken as 0.202 (g/cm^3) (Steele 1956; Setoyama et al. 1996). An effective adsorbent porosity of 68% and adsorption isotherms similar to Chemviron SC2 (Hauer and Day 2002) were also assumed. The resulting rough values at 10 bar were established to guide the early design of the regenerator plates and their values were not expected to be very precise.

The measured values are also comparable to the reported volumetric specific heat of adsorbed helium in aerogel, which is 0.58 J/cm^3-K at 5.25K (Wong and Chan 1990). The pore size in their aerogel sample ranges from 20 Å to 2000 Å, and the reported helium density in the aerogel is about 0.08g/cm^3.

The specific heat measurement uncertainty resulting from instrumentation accuracy and data acquisition system resolution is relatively small, less than approximately 1.5%. The scattering of the test data is mainly caused by the uncertainty in determining the initial breakthrough point based on the recorded temperature profiles. The uncertainty is mainly due to the high temperature measurement noise caused by using low excitation current to avoid sensor self-heating, as well as small regenerator temperature drifts due to parasitic heating.

SUMMARY AND FUTURE WORK

A practical design and fabrication approach to use the sensible heat of adsorbed helium to enhance the heat capacity of a low-temperature regenerator has been developed. The resulting adsorbent is an integral part of the heat spreader and is in intimate thermal contact with convective transfer surfaces. This fabrication approach also avoids potential particulate contamination issues in the regenerator associated with use of loose adsorbent particles. Experimental results over the temperature range 10 to 20 K show that the volumetric specific heat of the adsorbent/adsorbed helium is comparable to typical rare earth materials used in low temperature regenerators. The adsorbent regenerator technology has strong potential to significantly enhance the performance of low-temperature regenerative cryocoolers.

Future research in this area should include assembling a Stirling or Pulse Tube cryocooler with the microchannel helium adsorbent regenerator, characterizing its thermal and fluid performance, and assessing its robustness for potential tactical and space applications. In addition to low temperature regenerators, the monolithic adsorbent technology also has applications in cryo-sorption pumps, sorption coolers, and sorption compressors for JT coolers.

ACKNOWLEDGMENT

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